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Convective ripening and initiation of rainfall

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Abstract – This paper discusses the evolution of the droplet size distribution for a liquid-in-gas aerosol contained in a Rayleigh-Bénard cell. It introduces a non-collisional model for broadening the droplet size distribution, termed "convective ripening". The paper also considers the initiation of rainfall from ice-free cumulus clouds. It is argued that while collisional mechanisms cannot explain the production of rain from clouds with water droplet diameters of 20 μm, the non-collisional convective ripening mechanism gives a much faster route to increasing the size of the small fraction of droplets that grow into raindrops.

Introduction. – The dynamics of the onset of rainfall from ice-free ("warm") cumulus clouds is poorly understood [1–4]. Coalescence of droplets which collide due to differential rates of gravitational settling is effective for droplets with radius a above 50 μm, and leads to a runaway growth to produce millimetre-scale raindrops [5]. Many clouds are found to contain droplets with radius of approximately 8–15 μm [1–4], which result from primary condensation onto aerosol nuclei. For droplets in this size range, growth by collisional coalescence is slow because the product of the collision rate and the coalescence efficiency is low [1]. This makes it difficult to explain observations of the rapid onset of rainfall from warm cumulus clouds. (Rainfall from ice-bearing clouds is easier to explain: see [1] for a discussion of the Bergeron process.)

It is, therefore, desirable to formulate models for non-collisional growth of water droplets, in which some droplets are able to grow at the expense of others shrinking, by transferring water molecules between droplets as water vapour. Ostwald ripening [6] is one such mechanism, but it is too slow to be significant in terrestrial clouds [7,8], while it is relevant to test-tube models for rainfall [9]. It has been suggested that condensation processes may be able to cause the droplet size distribution to broaden due to fluctuations in the degree of supersaturation. This possibility has been addressed by numerous authors: see, for example, [10–18]. These investigations have used numerical simulations, and it is difficult to draw conclusions which are applicable to real clouds because of the limited range of size scales which can be simulated reliably. A difficulty with most of these models is that the droplets both grow and shrink as the supersaturation fluctuates. This work, however, introduces a model for which there is an asymmetry between growth and shrinkage.

This paper introduces a benchmark model for the broadening of the droplet size distribution of an aerosol due to convection. This process will be termed "convective ripening" to distinguish it from Ostwald ripening. This work considers how the process works in the simplest relevant model, which is an aerosol in a Rayleigh-Bénard convection cell. As well as having fewer physical parameters than a cloud, this system can be subject to a carefully controlled laboratory investigation.

Having described a non-collisional model for droplet growth, this will be applied to rain initiation from ice-free ("warm") cumulus clouds. An important aspect of this problem is that the conversion of a microscopic water droplet into a rain droplet is a very rare event (this fact was previously emphasised by Kostinski and Shaw [5]). However, the growth of a droplet to the stage where runaway growth occurs is a multi-stage process. The probability for the required number of collisions is extremely small. It is shown that droplets can grow much more rapidly by the convective ripening mechanism.

Discussions of convection processes within clouds often involve the complex and poorly understood issue of "entrainment" of air into a cloud (see, for example, [14,18]). This paper argues that a Rayleigh-Bénard cell appears to be a sufficient model to understand rainfall from warm

cumulus clouds, implying that hypotheses about entrainment do not appear to be necessary.

The convection cell model. – Consider a Rayleigh-Bénard convection cell, in which the working fluid is a gas (air, say) containing an aerosol suspension of liquid droplets (water, say). The height of the cell is h and the temperature difference between the upper and lower plates is ΔT_h. It is assumed that the horizontal dimensions of the cell are large compared to h. The gas has volume-specific heat capacity at constant pressure $C_{\rm g}$, density $\rho_{\rm g}$, kinematic viscosity ν and thermal diffusivity is $D_{\rm th}$. It will be assumed the convection in the container is in a turbulent regime, with rate of dissipation per unit mass ϵ.

The cell contains an aerosol of liquid droplets with density $\rho_{\rm l}$ and volume-specific heat of evaporation L. The vapour of the aerosol liquid in the carrier gas has diffusivity D. The number density of droplets is n_0, and the probability density function for the droplet radius a at time t is $P(a,t)$.

The objective is to understand how convection affects the distribution of sizes of the aerosol droplets. It will be assumed that the rate of collisions between the droplets is negligible. It will also be assumed that collisions of aerosol droplets with the walls of the container is not a significant process. The validity of this assumption is not critical to using this system as a model for cloud physics (because there is no material container in that context).

In order to understand the ripening of the droplet size distribution it is necessary to consider first how the aerosols responds to changes in the temperature of the surrounding gas, and then how the convection process influences the temperature.

Response to temperature fluctuations. – Changes in the temperature T of the surrounding gas cause the size of the droplets to change due to condensation or evaporation. This is characterised by two parameters, T_0 and $\tau_{\rm eq}$, which describe, respectively, the sensitivity and the timescale of the response. It will be shown that if the majority of the aerosol droplets have radius close to a_0, the change δa_0 of the equilibrium radius in response to a temperature increment δT satisfies

$$\frac{\delta a_0}{a_0} = -\frac{\delta T}{T_0} \tag{1}$$

to leading order in δT, and that the change in droplet radius occurs on a timescale $\tau_{\rm eq}$. In the following expressions for both T_0 and $\tau_{\rm eq}$ are obtained. Equivalent calculations can be found in many earlier works (reviewed in [1–4], see for example, [8,18]), but with differences in physical motivation and notation. A brief derivation is given here to make this paper unambiguous and self-contained.

The volume fraction of water molecules in the air, Φ, may be assumed to be uniform throughout the container because the system is well mixed by convection. This is the sum of contributions from water in the liquid and the vapour phase:

$$\Phi = \Phi_{\rm l} + \Phi_{\rm v}. \tag{2}$$

The equilibrium vapour content above a flat liquid surface at temperature T is denoted by $\Phi_{\rm eq}(T)$, and there may be a degree of supersaturation, denoted by s. It is assumed that the droplets are sufficiently large that curvature and hygroscopic effects of the aerosol condensation nuclei can be neglected. The volume fractions of water in the vapour and liquid phases are written in the form

$$\Phi_{\rm v} = \Phi_{\rm eq}(T) + s, \qquad \Phi_{\rm l} = \frac{4\pi}{3} n_0 \langle a^3 \rangle \tag{3}$$

(throughout this paper $\langle X \rangle$ denotes the expectation value of any quantity X). If the temperature of the gas changes, the sizes of the droplets will change due to condensation or evaporation. The rate of the condensation process is determined by diffusion of vapour. The radius of a droplet changes at a rate

$$\frac{{\rm d}a}{{\rm d}t} = j_{\rm v} = -\frac{D}{a}\Delta\Phi_{\rm v}, \tag{4}$$

where $j_{\rm v}$ is the volume flux density of condensing molecules and $\Delta\Phi_{\rm v}$ is the volume fraction on the surface of the droplet minus the volume fraction in the bulk of the gas phase. The surface of the droplet is in quasi-static equilibrium with the surrounding fluid, so there is no supersaturation at the surface. However, the temperature of the liquid droplet may be increased by an amount ΔT due to the latent heat of water condensing on the surface, so that

$$\Delta\Phi_{\rm v} = \frac{{\rm d}\Phi_{\rm eq}}{{\rm d}T}\Delta T - s. \tag{5}$$

The thermal flux density due to the latent heat is

$$j_{\rm th} = -L\frac{{\rm d}a}{{\rm d}t} = -\frac{D_{\rm th}C_{\rm g}}{a}\Delta T. \tag{6}$$

Combining (5) and (6) gives a simple expression relating the rate of droplet growth to the supersaturation

$$\frac{{\rm d}a}{{\rm d}t} = \frac{D_{\rm eff}}{a}s, \tag{7}$$

where the effective diffusion constant is

$$D_{\rm eff} = \frac{D}{1+\Theta}, \qquad \Theta = \frac{DL}{D_{\rm th}C_{\rm g}}\frac{{\rm d}\Phi_{\rm eq}}{{\rm d}T}. \tag{8}$$

This treatment neglected the possibility of cross-coupling between thermal and mass fluxes (the Soret effect). This could be incorporated with a simple modification of the theory, but the coefficients of the off-diagonal terms of the transport matrix do not appear to have been definitively determined.

In the case where the temperature of the system varies extremely slowly, the supersaturation is always negligible, and the relation between droplet size and temperature is determined by writing $\Phi = 4\pi n_0 a_0^3(T)/3 + \Phi_{\rm eq}(T)$ so that

a small change in temperature δT results in a small change of radius δa_0 given by (1), with coefficient

$$T_0 = 3\Phi_{\rm l}\left(\frac{{\rm d}\Phi_{\rm eq}}{{\rm d}T}\right)^{-1}. \tag{9}$$

Now consider the effect of varying the temperature of the aerosol at a finite rate, but still assuming that it is spatially homogeneous: write $T = T_0 + \delta T(t)$, $a = a_0(T_0) + \delta a(t)$, so that

$$\Phi = \frac{4\pi}{3} n_0 (a_0 + \delta a)^3 + \Phi_{\rm eq}(T_0) + \frac{{\rm d}\Phi_{\rm eq}}{{\rm d}T}\delta T(t) + s. \tag{10}$$

Taking the leading order in the small fluctuation δa and using (7) yields the following equation for the response of the droplets to fluctuations in temperature:

$$\frac{{\rm d}\delta a}{{\rm d}t} = -\frac{1}{\tau_{\rm eq}}\delta a - \frac{a_0}{\tau_{\rm eq} T_0}\delta T(t), \tag{11}$$

where the relaxation time is

$$\tau_{\rm eq} = \frac{a_0^2}{3\Phi_{\rm l} D_{\rm eff}} = \frac{1}{4\pi n_0 a_0 D_{\rm eff}}. \tag{12}$$

Now consider how to estimate the parameters T_0 and $\tau_{\rm eq}$. The rate of change of the saturation volume fraction is obtained from the Clausius-Clapeyron relation, ${\rm d}p/{\rm d}T = L/T\Delta V$, where ΔV is the volume change on a phase transition. Assuming that the vapour pressure is sufficiently low that the ideal gas law is applicable,

$$p = \frac{RT}{V_{\rm m}}\Phi_{\rm eq}, \tag{13}$$

where $V_{\rm m}$ is the molar volume of the liquid. Also, the volume change per mole associated with the phase transition is $\Delta V = V_{\rm m}/\Phi_{\rm eq}$. The Clausius-Clapeyron equation can therefore be written in the form

$$\frac{{\rm d}p}{{\rm d}T} = \frac{L}{T}\Phi_{\rm eq}. \tag{14}$$

Comparing (13) and (14) yields an expression for ${\rm d}\Phi_{\rm eq}/{\rm d}T$, and hence

$$T_0 = \frac{3RT}{LV_{\rm m} - RT}\frac{\Phi_{\rm l}}{\Phi_{\rm eq}}T. \tag{15}$$

For water at $T = 278\,{\rm K}$, $L = 2.4\times10^9\,{\rm J m^{-3}}$, $C_{\rm g} = 800\,{\rm J m^{-3}}$, $V_{\rm m} = 1.8\times10^{-5}\,{\rm m^3 mol^{-1}}$, $\Phi_{\rm eq} = 7\times10^{-6}$, $D = 2.5\times10^{-5}\,{\rm m^2 s^{-1}}$ and $D_{\rm th} = 1.9\times10^{-5}\,{\rm m^2 s^{-1}}$ [1,19]. These data yield $\Theta \approx 1$. If the liquid water content is 10% of the total water content, then $T_0 \approx 5\,{\rm K}$: that is, the droplet size is very sensitive to changes of temperature. If the droplets are of size $a = 10\,\mu{\rm m}$ and density $n_0 = 4\times10^8\,{\rm m^{-3}}$ (which are typical values for clouds [1,2,4]), the equilibration time is $\tau_{\rm eq} = 1.6\,{\rm s}$.

Ripening in a turbulent convection cell. – Now consider the response of the aerosol to convective motion in the cell. This is a consequence of how the temperature changes along the trajectories of the aerosol droplets (which are assumed to be advected by the flow).

Turbulent convection in a Rayleigh-Bénard cell is reviewed in [20–22]. The upper and lower plates are at temperatures $T_{\rm up}$ and $T_{\rm low}$, respectively. The expectation value of the temperature is close to $T_{\rm av} = (T_{\rm up} + T_{\rm low})/2$ (with small logarithmic corrections) [23], except in the vicinity of the upper and lower plates, and at any time most of the gas in the convection cell is at a temperature close to $T_{\rm av}$. Gas which is in contact with the lower plate of the cell is heated to a temperature $T_{\rm av} + \Delta T$ (where $0 \le \Delta T \le \Delta T_h/2$), and joins a plume of rising gas. The plumes are mixed by the turbulence in the interior of the cell. The plumes form fronts and later tendrils of approximately homogenous gas, which remain at a temperature close to the temperature that they had upon separation from the top or bottom plate until the last stage of the mixing process. In the final stage of mixing a tendril formed by the plume mixes rapidly with gas from the interior of the cell, which is at a temperature close to $T_{\rm av}$. The final stage of mixing occurs very rapidly, on the Kolmogorov timescale, $\tau_{\rm K} = \sqrt{\nu/\epsilon}$. The time $\tau_{\rm mix}$ between a plume separating and the gas surrounding a given aerosol particle becoming mixed is highly variable: $\tau_h \gg \tau_{\rm mix} \gg \tau_{\rm K}$, where $\tau_h = (h^2/\epsilon)^{1/3}$ is the turnover time of the largest eddies. The equilibration timescale will be assumed to lie between the timescales describing the flow: $\tau_{\rm mix} \gg \tau_{\rm eq} \gg \tau_{\rm K}$.

The consequence of this picture is that droplets in a rising plume are at a temperature, $T_{\rm av} + \Delta T$, and while the plume forms they equilibrate to a smaller radius: to leading order

$$a_- = a_0 - \Lambda\Delta T, \qquad \Lambda \equiv \frac{a_0}{T_0}. \tag{16}$$

The gas in the plume rises, without cooling due to heat exchange, until it reaches the interior of the cell. After a timescale $\tau_{\rm mix}$, the gas in the plume starts to mix with the gas in the interior. This mixing happens on a timescale which is short compared to the phase equilibration time, so that droplets of size a_- are mixed with the droplets in the bulk, which are of size a_0. Similarly, plumes of cold gas which form on the upper plate at a temperature $T_{\rm av} - \Delta T$ inject larger droplets, of radius $a_+ = a_0 + \Lambda\Delta T$. The final stage of this mixing happens on a timescale of the Kolmogorov time $\tau_{\rm K}$, which is small compared to the time $\tau_{\rm eq}$ required for aerosol droplets to come into equilibrium. It follows that while the temperature fluctuations associated with the plume are dissipated, fluctuations in the droplet size remain "frozen in", resulting in a broadening of the droplet size distribution.

Now consider how this model is used to model the evolution of the droplet size distribution, $P(a,t)$. The plumes carrying gas away from the lower plate have a distribution of temperature. Let $J(\Delta T)\,\delta\Delta T$ be the volume of gas per unit area, per unit time, which rises from the lower plate and which has a temperature $T_{\rm av} + \Delta T$, with ΔT in

the interval $[\Delta T, \Delta T + \delta\Delta T]$. It is assumed that the flux from the upper plate may be described by the same function $J(\Delta T)$, where the temperature of the falling plume is $T_{\rm av} - \Delta T$. The material in this small temperature range $\delta\Delta T$ occupies a volume fraction of the gas in the column which increases at a rate

$$\frac{1}{V}\frac{{\rm d}V}{{\rm d}t} = \frac{J(\Delta T)}{h}\delta\Delta T. \tag{17}$$

The droplets in this volume fraction undergo a change of radius equal to $\Delta a = -\Lambda\Delta T$. Taking account of contributions from the upper and lower plates, this results in a change of the droplet size distribution which satisfies

$$\frac{\partial P}{\partial t}(a,t) = \int_0^\infty {\rm d}a'\ \mathcal{K}(a,a')\,P(a',t), \tag{18}$$

where the kernel may be approximated by $\mathcal{K}(a,a') = K(a-a')$ with

$$K(\Delta a) = \frac{1}{\Lambda h}[J(\Delta a/\Lambda) + J(-\Delta a/\Lambda)] - \frac{1}{\tau_{\rm r}}\delta(\Delta a). \tag{19}$$

Here the delta function ensures conservation of particle number, and $\tau_{\rm r}$ is an estimate for the timescale of a convection roll:

$$\frac{1}{\tau_{\rm r}} = \frac{2}{h}\int_0^\infty {\rm d}x\ J(x). \tag{20}$$

The time $\tau_{\rm r}$ cannot exceed the integral timescale of the flow: $\tau_{\rm r} \le \tau_h = (h^2/\epsilon)^{1/3}$.

The change in droplet radius arises from the abrupt change in temperature at the hot and cold plates of the convection cell. The droplets change size discontinuously, with a magnitude comparable to $\delta a_{\rm max} = \Lambda\Delta T_h/2$. Consider the evolution of a droplet which spends some time close to the top of the cell and which has repeated encounters with the cold plate. As its temperature rises and falls, we might expect that the radius of the droplet would increase and decrease, with a minimal net effect. However, there can be a marked asymmetry between the timescales of heating and cooling. In a convection cell the gas is cooled relatively slowly by the upper plate, before being warmed rapidly by turbulent mixing when a plume falls into the interior. If the cooling occurs slowly compared to $\tau_{\rm eq}$, droplets grow by condensation. If the warming due to mixing is on a timescale $\tau_{\rm K} \ll \tau_{\rm eq}$, the larger droplets do not evaporate to their original size. The asymmetry between the cooling and heating processes allows the droplets to have a systematic growth, rather than a cyclic fluctuation of size.

The production of rain from clouds depends upon droplets reaching a size which is significantly larger than their original size. In the context of the Rayleigh-Bénard model, this would require a droplet to undergo repeated encounters with the cold plate. The time $\tau_{\rm mix}$ that an aerosol particle spends in a plume before it is mixed is highly variable and it will usually be very short compared to the integral time τ_h. It follows that some droplets may experience many interactions with the upper plate in rapid succession, as illustrated in fig. 1.

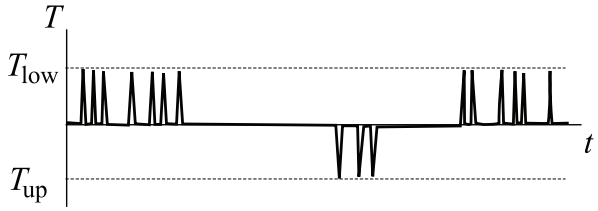

Fig. 1: Schematic plot of the temperature of a convected aerosol particle. The time $\tau_{\rm mix}$ between contacts with the upper plate may be as short as $\tau_{\rm K}$. There may be a very large number of contacts with the upper plate before the particle reaches the lower plate, which requires a much longer time, of order τ_h.

The problem of warm rain initiation. – A cloud contains water droplets formed by condensation onto microscopic nuclei such as salt granules, dust grains, or particles of organic matter. Their concentration and droplet radius are quite variable, but the remainder of this paper uses the following representative values [1–4] for a convecting cumulus cloud which could produce precipitation. The typical droplet radius is $a_0 = 10\,\mu$m, the number density is $n_0 = 4\times10^8\,{\rm m}^{-3}$, and the cloud depth is $h = 10^3\,$m. The rate of decrease of temperature with height (lapse rate) is $3\,^\circ$C per 1000 ft, which exceeds the adiabatic lapse rate by $1\,^\circ$C per 1000 ft, so that the effective temperature difference between the top and bottom of the cloud is $\approx 3\,$K. The typical vertical velocity of air inside the cloud has magnitude $2\,{\rm m\ s}^{-1}$, so that the eddy turnover time may be taken to be $\tau_h = 10^3\,$s [1,2]. An estimate for the rate of dissipation is $\epsilon \approx h^2/\tau_h^3 = 10^{-3}\,{\rm m}^2{\rm s}^{-3}$, which gives an estimate of the Kolmogorov time $\tau_{\rm K} \approx 10^{-1}\,$s. Rain falls as droplets of size approximately $a = 1\,$mm. A rate of rainfall of $3.6\,{\rm mm\ h}^{-1} = 10^{-6}\,{\rm m\ s}^{-1}$ is described as moderate to heavy rainfall [1].

The coalescence efficiencies ε of small droplets are somewhat uncertain, but it is widely accepted that they are low [1,2]. If the larger droplet has radius below $20\,\mu$m, it is believed that $\varepsilon \le 0.1$, and that for radius $10\,\mu$m, $\varepsilon \le 0.03$ [2]. For droplets of size $a = 50\,\mu$m colliding with droplets of size $a = 10\,\mu$m, however, the efficiencies are expected to be close to unity [1,2].

Collisions between droplets settling at a different rate yield a very small collision rate. The Stokes law for the drag on a sphere at low Reynolds number indicates that the gravitational settling rate is [1,2]

$$v = \tau_{\rm p} g = \kappa a^2, \qquad \kappa = \frac{2}{9}\frac{\rho_{\rm l}}{\rho_{\rm g}}\frac{g}{\nu}, \tag{21}$$

where $\tau_{\rm p}$ is the response time characterising the Stokes drag on a droplet. Inserting values for air and water at $5\,^\circ$C gives $\kappa \approx 1.4\times10^8\,{\rm m}^{-1}{\rm s}^{-1}$. The collision rate of a droplet of radius $a+\Delta a$ with a gas of particles of radius a is

$$\mathcal{R} = \pi\varepsilon n_0(2a+\Delta a)^2\kappa[(a+\Delta a)^2 - a^2] \sim 8\pi\kappa\varepsilon n_0 a^3\Delta a. \tag{22}$$

Setting $\Delta a = 2.5\,\mu$m and $\varepsilon = 0.03$ in addition to the parameters defined above gives $\mathcal{R} \approx 10^{-4}\,\mathrm{s}^{-1}$. The rate of coalescence of typical sized water droplets due to collisions is therefore very small.

Saffman and Turner [24] investigated the role of turbulence in facilitating collisions between water droplets. In the case of very small droplets, the collision rate due to turbulence is a consequence of shearing motion, so that the collision speed is of order a_0/τ_K. They argue that the corresponding collision rate is

$$\mathcal{R}_\mathrm{turb} = \sqrt{\frac{8\pi}{15}}\frac{n_0\varepsilon(2a)^3}{\tau_\mathrm{K}}. \tag{23}$$

For the parameters of the cloud model, the gives $\mathcal{R}_\mathrm{turb} \approx 2\times10^{-6}\,\mathrm{s}^{-1}$, which is negligible. The effects of turbulence are dramatically increased when the effects of droplet inertia are significant. Inertial effects are measured by the Stokes number, $\mathrm{St} \equiv \tau_\mathrm{p}/\tau_\mathrm{K}$. The collision rate is greatly enhanced by effects due to caustics for $\mathrm{St} > 0.3$ [25], but the parameters of our cloud model yield $\mathrm{St} \approx 10^{-2}$, where there is no significant enhancement.

After a droplet has grown to a size where it is much larger than the typical droplets, and where the coalescence efficiency is approximately unity, it falls rapidly and collects other droplets in its path. Consider a droplet of size a_1 falling through a "gas" of small droplets, which can be characterised by the liquid volume fraction $\Phi_\mathrm{l} = 4\pi n_0\langle a^3\rangle/3$. The large droplet falls with velocity $v = \kappa a_1^2$ and grows in volume at a rate $\pi a_1^2\Phi_\mathrm{l} v$, so that

$$\frac{\mathrm{d}a_1}{\mathrm{d}t} = \frac{\kappa\Phi_\mathrm{l}a_1^2}{4}. \tag{24}$$

Solving this equation shows that the droplet radius diverges in the time

$$\tau_\mathrm{div} = \frac{4}{\kappa\Phi_\mathrm{l}a_1}. \tag{25}$$

For the model parameters, a droplet of size $a_1 = 50\,\mu$m requires time $\tau_\mathrm{div} \approx 2\times10^3\,\mathrm{s}$ to undergo divergent growth. According to (25) the time before runaway growth is expected to occur increases rapidly as the droplet size gets smaller, and this estimate must be a lower bound because it ignores the effects of collision efficiency and the settling velocity of the smaller droplets.

Rare events and rain initiation. – Consider the rate at which droplets must reach the size threshold for runaway growth. Rainfall at a rate of $3.6\,\mathrm{mm\,h}^{-1} = 10^{-6}\,\mathrm{m\,s}^{-1}$ is considered as "moderate". If the raindrops have size $a \approx 1\,\mathrm{mm}$, this corresponds to raindrops falling at a rate of approximately $250\,\mathrm{m}^{-2}\mathrm{s}^{-1}$. Given the assumed cloud depth of $h = 10^3\,\mathrm{m}$, the volumetric rate of production of raindrops is approximately $0.25\,\mathrm{m}^{-3}\mathrm{s}^{-1}$. If the microscopic droplets have density $n_0 = 4\times10^8\,\mathrm{m}^{-3}$, then the rate of conversion of each microscopic droplet into a "collector" droplet undergoing runaway growth is approximately $6\times10^{-10}\,\mathrm{s}^{-1}$. An alternative statement is that if a shower lasts for five minutes, the probability that any given water droplet has grown to become a rain droplet is small, approximately 2×10^{-7}. The problem of rain initiation is, therefore, concerned with the frequency of very rare events. This point has also been made by Kostinski and Shaw [5].

Despite the fact that the required conversion probability is very small (of order 10^{-7}), growth of droplets is too slow by a collisional mechanism. On growing from $10\,\mu$m to $50\,\mu$m, the volume of a droplet increases by a factor of 125, that is, there are roughly 100 collision events. It was argued above that the rate for the first collision events is small, $\mathcal{R}_0 \approx 10^{-4}\,\mathrm{s}^{-1}$. Even allowing for the fact that the collision rates increase as the droplet grows, the probability for the obtaining 100 collisions after $t = 10^3\,\mathrm{s}$ will be much smaller than 10^{-7}.

Fast droplet growth by the convective mechanism. – As well as the theoretical difficulties of explaining droplet growth by collisional processes, observational evidence is difficult to reconcile with a collisional mechanism. Clouds may exist for long periods, before quite suddenly producing rainfall. The rapid onset of rainfall is usually associated with convective instability, which (because of the large Reynolds number) implies turbulent motion. Equation (23) indicates that the role of turbulence in facilitating particle collisions is negligible for the small droplets in the model treated here, implying that the rainfall is triggered by some other aspect of the convective process. If a parcel of air is lifted by convection, condensation occurs as the temperature falls. However, the fractional increase of the droplet size which can be achieved is not large enough to start runaway growth, and increasing the size of droplets by condensation also reduces the dispersion of their radius.

For these reasons it is desirable to find other mechanisms whereby convection can result in runaway growth. The Rayleigh-Bénard cell can serve as a model for convective motion in a cloud, and it will be argued that the convective ripening mechanism can result in droplet growth rates which are more rapid than the collisional mechanism.

Droplets can grow or shrink due to changes in the level of supersaturation. Consider the circulation of a droplet in a rising packet of air. This air mass is cooled *slowly* (on a timescale which is much larger than τ_eq) by radiation when it reaches the top of the cloud. The droplets that it contains increase in size by condensation, due to capacity of the surrounding air to carry water vapour being reduced. The cool packet of air then becomes part of a "plume" of descending air, which falls far into the body of the cloud before being *rapidly* mixed with ambient air (on a timescale $\tau_\mathrm{K} \ll \tau_\mathrm{eq}$). The mixing occurs so rapidly that the droplets are unable to evaporate, and their increased sizes are "frozen". Note that the asymmetry between a slow cooling and a rapid warming ensures that the mass gained by condensation is not lost to

evaporation. The mechanism requires $\tau_{\rm eq} \gg \tau_{\rm K}$. This inequality is easily satisfied for the model cloud parameters, where $\tau_{\rm eq} \approx 1\,{\rm s}$ and $\tau_{\rm K} \approx 10^{-1}\,{\rm s}$.

The temperature of the descending plume increases due to adiabatic compression as the air pressure rises, but it is still colder than the surrounding gas at the time when mixing occurs.

Despite the characteristic temperature T_0 being quite small, one single cycle of this process is not sufficient to bridge the bottleneck and achieve runaway growth. Some aerosol droplets will make a large number of contacts with the upper surface of the cloud, increasing their size on each cycle, and it is proposed that these droplets become the collectors which undergo runaway growth. Only a very small proportion ($\approx 10^{-7}$) of aerosol droplets have to reach the required size. The time for an aerosol particle at the top of the cloud to join a descending plume, undergo mixing, and then to be advected back to the upper surface of the cloud is highly variable. The time between these growth events can be as short as few multiples of $\tau_{\rm eq}$. This timescale is typically less than 10 s, which is much smaller than the timescale between collisional coalescence of small droplets (typically $10^3\,{\rm s}$). These considerations show that the convective ripening mechanism is a strong candidate for creating "collector" droplets.

Equation (7) implies that growth of water droplets by condensation increases the area of a droplet by an amount which is independent of the droplet size on each cycle. This means that as well as having a faster attempt rate, the convective ripening mechanism also has the advantage that the radius increases by a factor of $N^{1/2}$ after N steps, instead of $N^{1/3}$ for the collisional process.

Conclusion. – This paper has described a non-collisional model for increasing the dispersion of droplet sizes in a Rayleigh-Bénard cell. This leads to a proposed resolution of the droplet growth bottleneck problem in cloud physics. It has been argued above that the dominant mechanism for creating larger droplets is that droplets grow by condensation at the cold upper surface of a cloud, but that the increased size is frozen in when a falling plume of cold air is mixed rapidly in the interior of the cloud.

The convective ripening mechanism was compared with collisional growth in clouds. Both processes require many growth events to produce a rare collector droplet which undergoes runaway growth. The convective ripening mechanism discussed here allows the growth steps to occur at a much higher rate. Further work in required to quantify the statistics of repeated contacts with the cool plate of the convection cell, because this determines the rate at which the largest droplets can grow.

This paper has presented a benchmark model for the growth of aerosol droplets by condensation. Real clouds are extremely complex structures with a substantial number of competing physical processes. The temperature profile and the mechanism of cooling at the upper surface are very different from a Rayleigh-Bénard cell. It will be a useful exercise to assess the extent to which this model can be developed to provide a quantitative description of real clouds.

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